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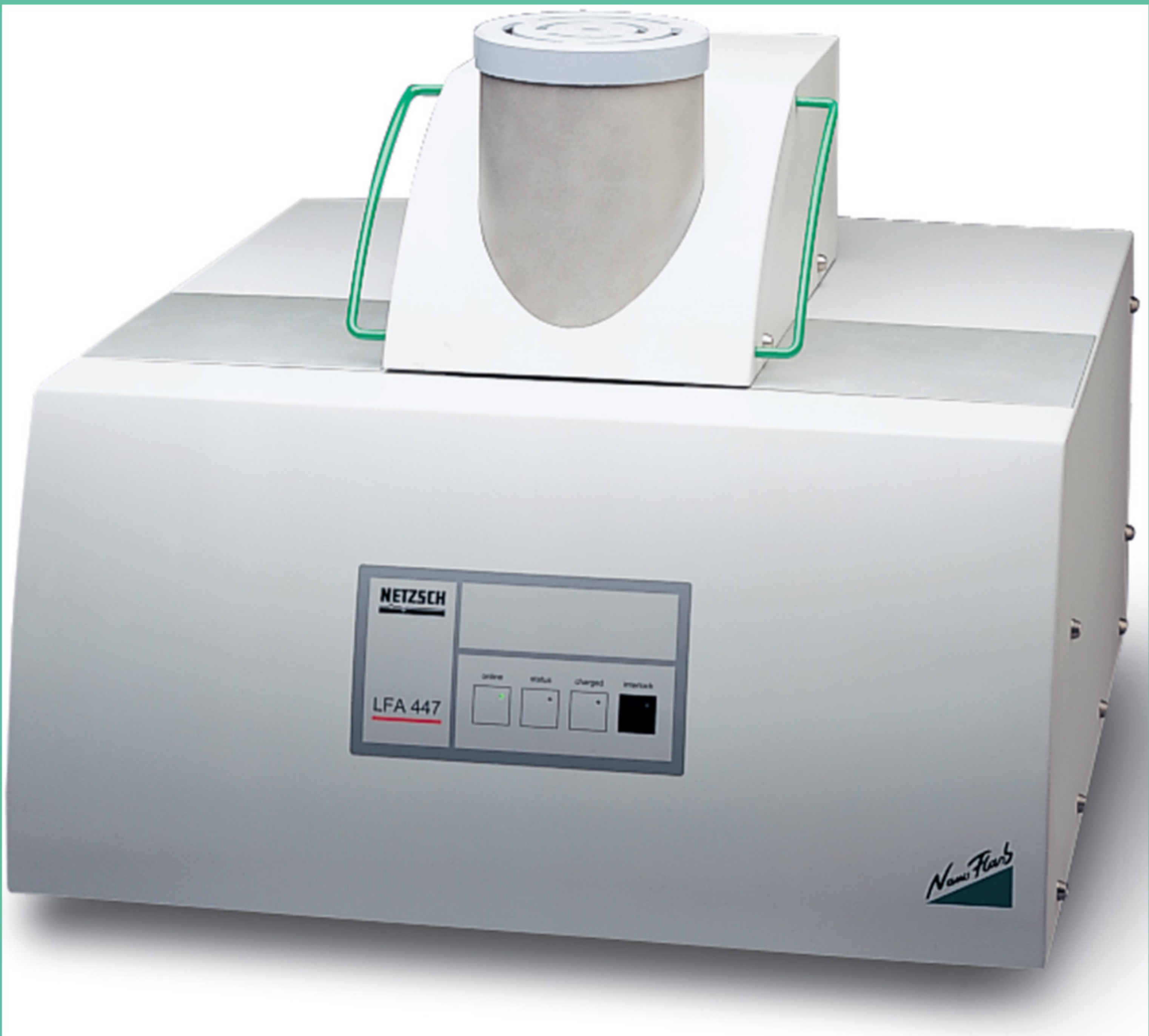

COATING TRANSLUCENT AND SEMITRANSPARENT MATERIAL SAMPLES FOR LASER FLASH ANALYSIS

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Contents

1. Foreword

The aim of this technical report is to present and discuss the influence of graphite coating on the measurement of thermal diffusivity for translucent or semitransparent material samples with the Laser Flash Analysis (LFA) method [1]. This experimental study has been conducted at the Building Material Characterization Laboratory of Aalborg University - Department of the Built Environment [2], with the Laser Flash Apparatus LFA 447 (Netzsch Gerätebau GmbH) [3].

2. Introduction and problem statement

The LFA method consists in shining a laser flash pulse on one side of the tested sample and record the temperature change as a function of time on the other side of the sample with an infrared (IR) detector. The thermal diffusivity is then calculated from this temperature signal record [1].

The key of a proper LFA measurement resides in getting a reliable surface temperature measurement with the instrument's IR sensor and having the laser pulse totally and homogeneously absorbed by the surface of the tested sample without penetration of the former in the bulk of the sample. The sample must thus fulfil the following important criteria [4]:

- The sample must not be translucent or transparent in the visible light and near-IR.
- The sample must not reflect light.
- The sample must present good surface emission and absorption.

Many common materials do not satisfy these aforementioned criteria. Many polymers and glasses are translucent or semitransparent in the visible and near-IR light. Therefore, the flash lamp laser pulse is not absorbed by the tested sample, it is transmitted through the materials and reaches directly the IR detector. The laser light saturates the IR detector and there is no useful measurement signal that can be used to calculate the thermal diffusivity of the tested sample.

On the other hand, metals are highly reflective, and thus do not absorb the laser pulse either. This is not an adequate situation for performing a proper LFA measurement.

Only a few materials, such as pure graphite, are non-reflective and opaque to the flash lamp laser. They can thus be used as is to perform LFA measurement. For all the other material, it is necessary to apply coating prior to the LFA measurement in order to ensure laser pulse absorption and reliable surface temperature measurement, and obtain a good signal with a high signal-to-signal ratio for the determination of the thermal diffusivity.

In general, all samples should be coated on both sides with a thin layer of a material that presents a high thermal conductivity (to not impact the overall thermal diffusivity of the test sample), a high absorbance of to the laser flash light, and a high emissivity (for the surface temperature measurement by the IR detector). It is the preferred method to improve the sample's emission/absorption surface properties and optimize the signal-to-noise ratio [4].

In the case of specific heat capacity measurement with an LFA apparatus (comparison method), it is absolutely necessary to coat both the test sample and the reference sample. The coating on both samples should be the same because this LFA comparison method requires that the test sample and the reference sample have the absorption and emissivity surface properties [4].

The two common coating technics for LFA test samples are gold coating and graphite coating. Graphite coating is the standard coating technic for LFA samples as it is very cheap, fast, and effective [4]. Graphite has high thermal conductivity (minimum disturbance to the overall thermal diffusivity of the test sample), a high absorption to the flash lamp laser pulse, and a high emissivity. A graphite spray is applied and dried several times on the sample to form a very thin graphite layer (around 5 µm).

However, for some specific types of test samples, the graphite coating is not appropriate: very thin, transparent samples where graphite layer can be too thick compared to the test sample; samples composed of a material that can potentially react with carbon (especially at high temperatures) such as steel [4]. For those cases, the gold coating method is then necessary to make the sample opaque to the laser pulse. An extremely thin gold layer (few nanometers) is deposited onto the sample's surface (gold sputter coating or sputter deposition). The gold coating is very efficient for good absorption of the laser pulse, but it is expensive (around 500 € for the gold sputter coating of one sample) and it requires special equipment. The gold-coated sample should then be dusted with graphite in order to increase its emissivity and absorptivity [4].

In this report, only translucent or semitransparent samples made of materials with "standard" thermal diffusivity (materials such as, e.g., ceramics with relatively low thermal diffusivity such that the LFA measurement duration is larger than 100 ms) and on which graphite coating can be applied are considered. The conclusions of this study can be extended to all samples that can be coated with graphite, expect for materials with very high thermal diffusivity for which the LFA measurement duration is smaller than 100 ms. Mehling et al. (1998) [5] have compared the accuracy of thermal diffusivity measurement with the LFA method when testing two semitransparent samples (microscope slide glass and high-grade fused silica platelet) coated with either of the two abovementioned standard coating technics. It was reported that the measurements on the gold-coated samples were very accurate. For the graphite coating, however, an initial long-wave direct radiative heat transfer ("ballistic transport") is observed inside the semitransparent test sample. In the case of gold coating, this direct radiative heat transfer is suppressed because of the low emissivity of the gold layer. But this is not the case for the graphite coating, and it leads to a systematic overestimation of the thermal diffusivity (up to 10% at 773 K) if it is not accounted in the LFA data analysis. To tackle this issue, Mehling et al. have developed and validated a new numerical fitting model to calculate accurately the thermal diffusivity of the semitransparent sample and compensating for this initial direct radiative heat transfer. This new model (denominated "Radiation" or "Radiation + pulse correction" model for the Netzsch LFA 447) thus accurately assesses the thermal diffusivity of semitransparent samples coated with graphite coating only. This is a major improvement as it spares the use of the expensive gold coating.

The current investigation aims to reproduce the results of Mehling et al. (1998) to ensure that the thermal diffusivity of semitransparent test samples can be accurately determined by the LFA method with graphite coating only and using the "Radiation" fitting model. This study also assesses the influence of different thicknesses for the graphite coating layer on the measurement results.

3. Materials and methodology

For this study, two translucent ceramic reference samples of known thermal diffusivity are chosen:

- The Pyrex 7740 (non-coated, semitransparent): Ø = 12.45 mm ; δ = 1.987 mm.
- The Pyroceram 9606 (gold-coated, opaque): Ø = 12.65 mm ; δ = 1.990 mm.

Both samples are tested at 25 °C and 200 °C with the LFA method to assess their thermal diffusivity with different graphite coating layer thicknesses (expressed in "number of layers" or %mass of the tested sample). The thermal diffusivity is calculated with three different fitting models: "Adiabatic + pulse correction"; "Cowan + pulse correction"; "Radiation + pulse correction" [1]. As mentioned before, the "Radiation + pulse correction" model is the fitting model developed by Mehling et al. (1998) to eliminate the thermal diffusivity overestimation due to the initial long-wave direct radiative heat transfer [5].

For each measurement (average of 20 individual LFA pulses) the error is calculated as the difference between the measured data and the stated thermal diffusivity of the reference sample at the specified temperature. Those "true" values (stated reference values) of thermal diffusivity for the reference samples have been accurately determined by Netzsch Gerätebau GmbH (manufacturer of the LFA apparatus and the reference samples [3]).

The Pyrex 7740 sample is used to verify that graphite coating can be used in combination of the "Radiation" fitting model for the accurate measurement of the thermal diffusivity of semitransparent materials. It also provides an indication of the optimum graphite layer thickness for the LFA tests.

The gold-coated Pyroceram 9606 sample is used to check if additional graphite coating on gold-coated samples has any influence on the accuracy of the thermal diffusivity measurement.

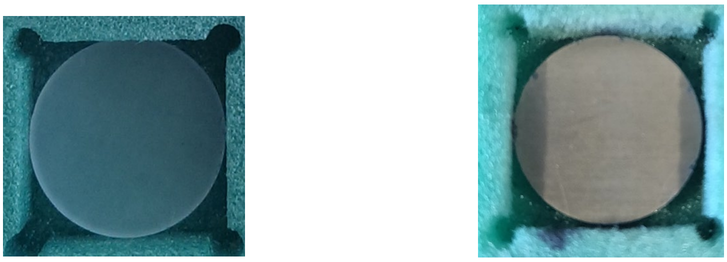

Figure 1: *LFA reference samples: Pyrex 7740 (left); Pyroceram 9606 (right).*

4. Results and discussion

4.1. Pyrex 7740: optimum layer thickness for graphite coating on a semitransparent sample

As explained above, the thermal diffusivity of the semitransparent reference sample Pyrex 7740 is measured with different layer thicknesses of graphite coating.

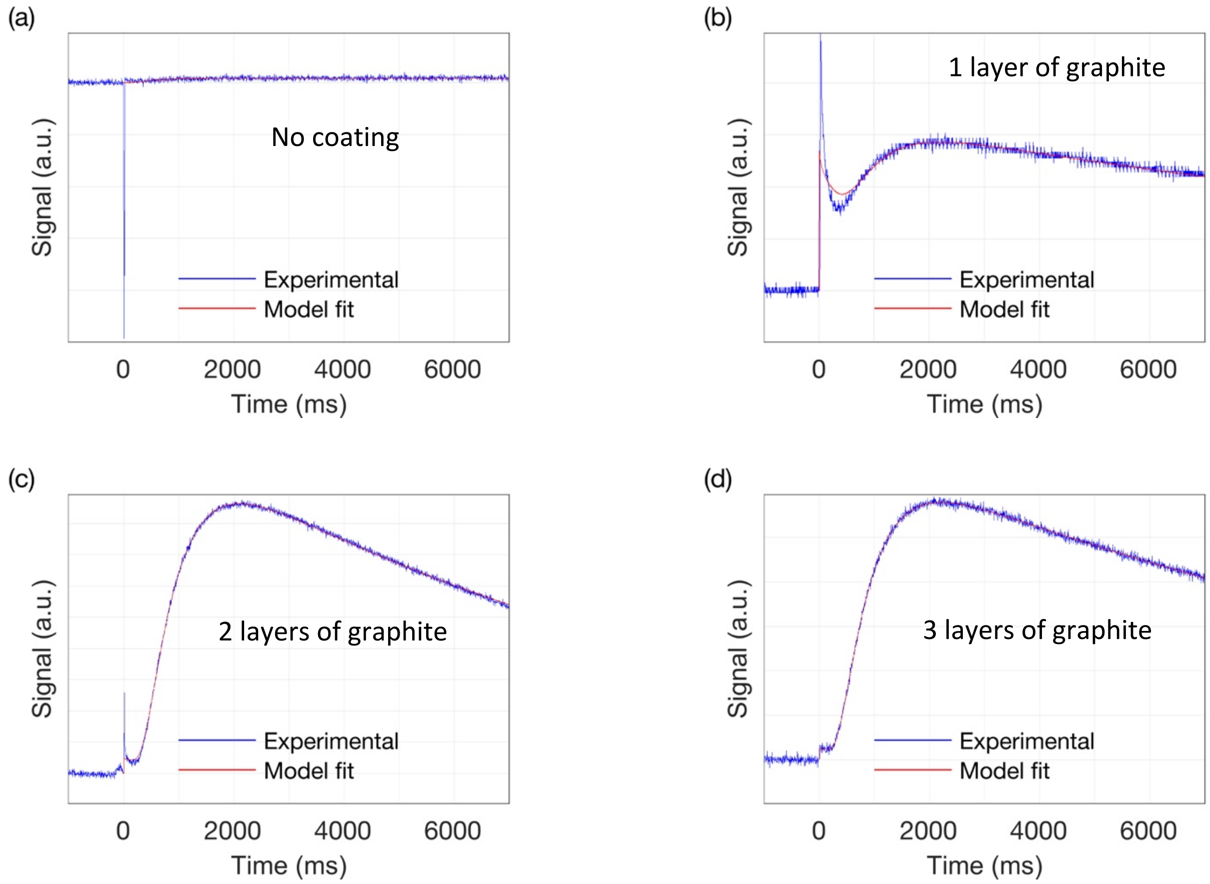

Figure 2: *LFA signals (surface temperature measure with IR detector as a function of time) for sampled of Pyrex 7740 with different thicknesses of graphite layers: (a) no coating, (b) 1 layer of graphite, (c) 2 layers of graphite, and (d) 3 layers of graphite.*

One can see in *Figure 2* the strong impact of the number of graphite layer on the quality of the LFA signal (used to calculate the thermal diffusivity with a fitting model). Without any coating on the semitransparent material sample (*Figure 2 (a)*) the light of the laser flash is not absorbed but transmitted directly through the sample. The laser pulse saturates the IR detector and there is no useful measurement signal that can be used to calculate the thermal diffusivity. With only one layer of graphite coating (*Figure 2 (b)*) there is insufficient absorption of the laser pulse. A large part of the laser pulse is directly transmitted through the semitransparent sample and induces a very large initial peak in the IR detector recordings. There is also a large initial direct radiative heat transfer inside the test sample due to the fact that a part of the laser pulse that is transmitted through the bulk of the semitransparent materials heats up directly the graphite layer on which the surface temperature recording by IR detector is performed. The initial signal peak and the direct radiation transmission are then followed a slower signal increase that is due to the thermal diffusion inside the bulk of the tested sample. Upon increasing the number of graphite coating layers on the semitransparent test sample, the laser flash light is better absorbed and diminishes or fully eliminates the initial peak of direct light transmission through the sample to the IR detector. With two layers of graphite coating (*Figure 2 (c)*) the initial direct light transmission is reduced to an acceptable extent. The laser pulse is almost entirely absorbed by the graphite coating of the surface shined by the laser, and the signal is adequate for determination of thermal diffusivity. For graphite coating of three or more layers (*Figure 2 (d)*) the laser pulse is entirely absorbed by the surface shined by the laser and the initial signal peak of direct light transmission is totally eliminated. The initial direct long-wave radiation heat transfer remains (although with a much lower intensity than when the laser pulse is not properly absorbed by the sample's surface) and is responsible for the initial step in the signal.

Based on those observations, it is recommended to apply 3 layers of graphite coating when performing LFA test on a semitransparent material.

In order to calculate the thermal conductivity of the test sample from the recorded LFA signal, different fitting models can be applied. There are three of these fitting models: the "Adiabatic + pulse correction" model assumes adiabatic conditions; the "Cowan + pulse correction" takes into account the heat losses by convection and radiation between the surface of the tested sample and its surrounding environment; the "Radiation + pulse correction" takes into account the heat losses of the sample and the initial direct radiative heat transfer ("ballistic transport") [1][5]. These models are tested here and compared to each other for the LFA tests of the Pyrex 7740 with different thicknesses of graphite coating.

One can see in *Figure 3* and *Figure 4* the thermal diffusivity measurement error for the reference sample Pyrex 7740 with different coating thicknesses at 25 °C.

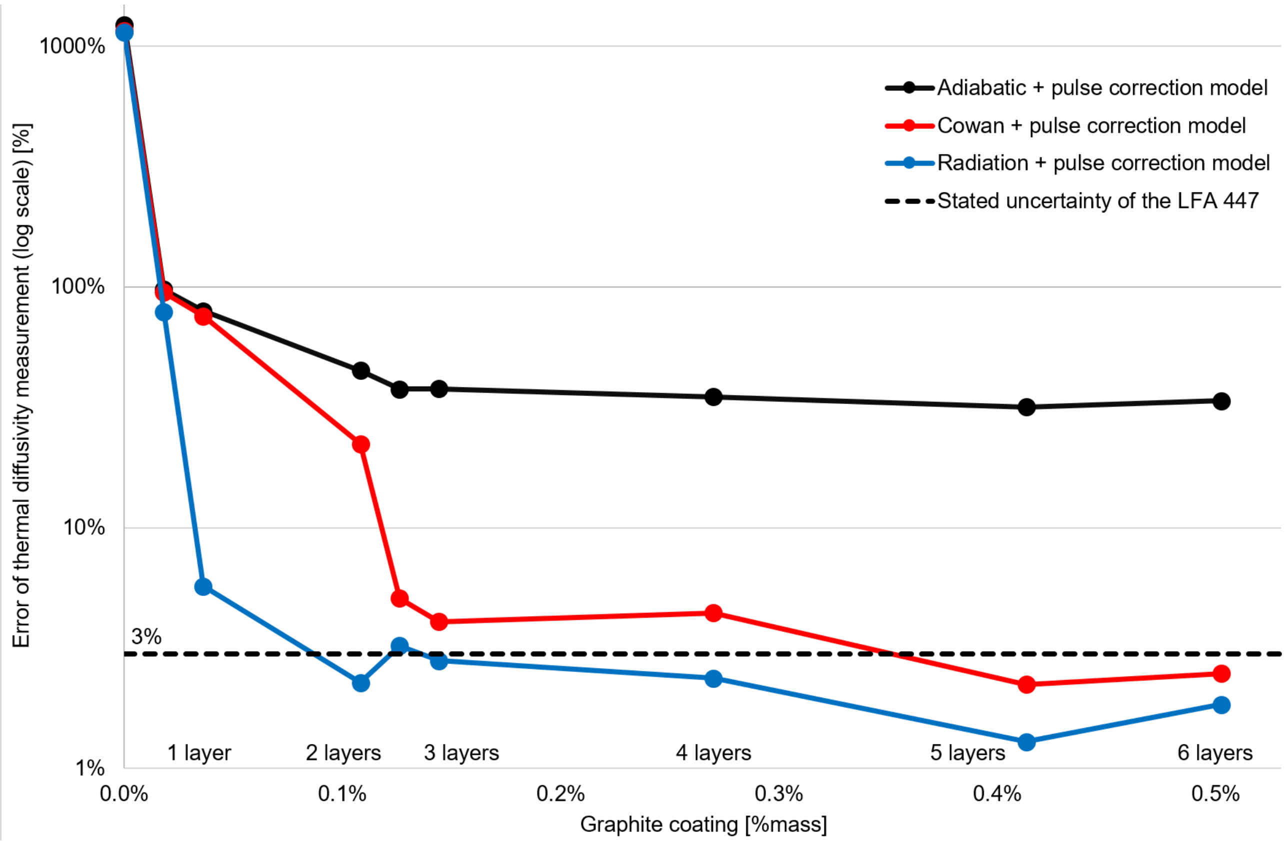

Figure 3: *Error of thermal diffusivity determination of the Pyrex 7740 (25 °C) as a function of numbers of graphite coating layers for three different fitting models.*

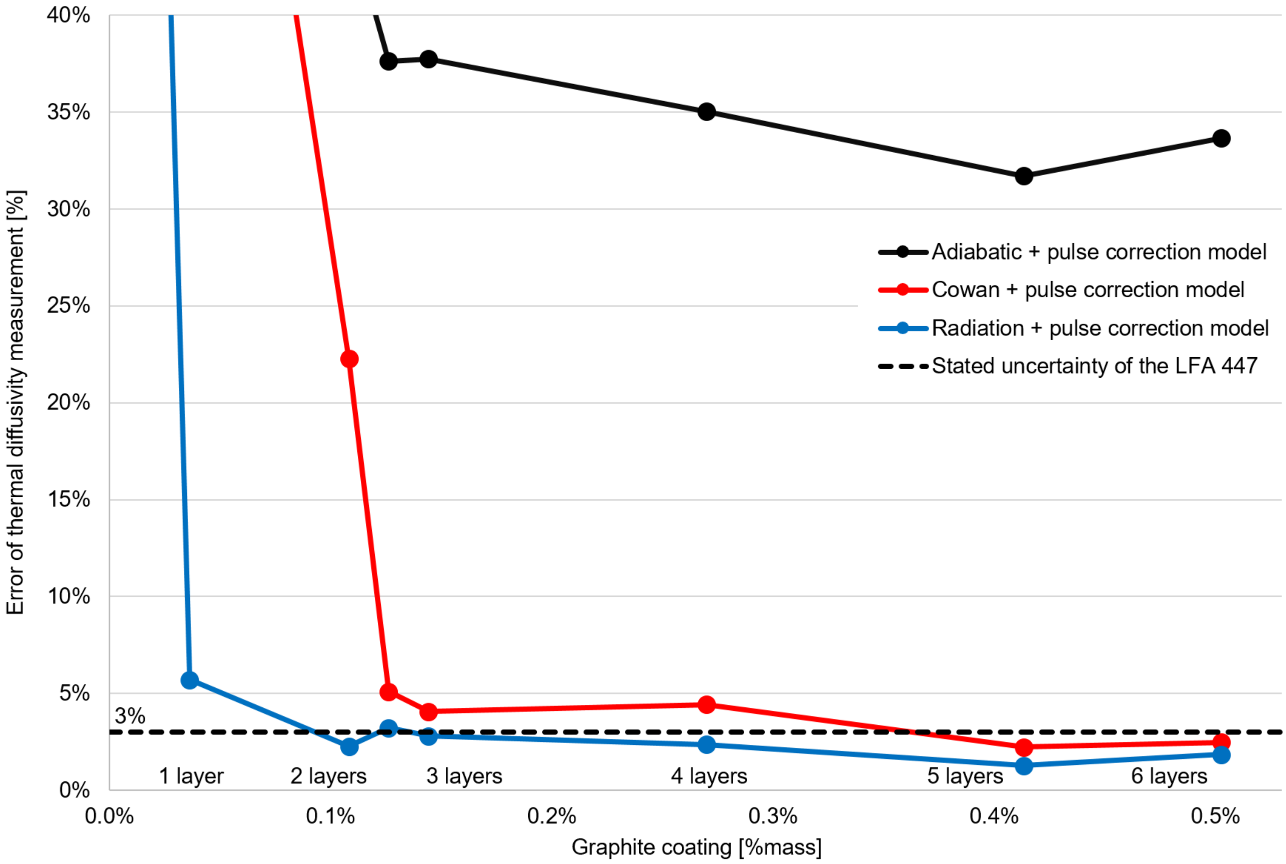

Figure 4: *Error of thermal diffusivity determination of the Pyrex 7740 (25 °C) as a function of numbers of graphite coating layers for three different fitting models.*

As expected, the aberrant measurement signals resulting from the LFA test with no or insufficient graphite coating (no coating, partial coating of half a layer, one layer only) produce erroneous thermal diffusivity results (>1000% of error). For graphite coating of two layers and more, the thermal diffusivity can be determined adequately. Graphite coatings thicker than three layers do not significantly change the error for the thermal diffusivity measurement. A large number of graphite coating layers (in here 6) does not significantly influence negatively the LFA measurement either. This is due to the fact that the additional thermal resistance of the graphite coating is negligeable in comparison to the thermal resistance of the test sample: the thickness of the graphite coating is negligeable in comparison to that of the sample, and the thermal conductivity of the graphite coating is much larger than that of the material of the test sample.

Regarding the different fitting models, the "Adiabatic + pulse correction" model performs very poorly, which is expected since the experimental conditions are far from being adiabatic (significant heat losses to the surrounding because of the relatively long duration of the LFA experiment due to the relatively low thermal diffusivity of the tested sample). These heat losses to the surrounding (convection and radiation) are accounted for by the "Cowan" and the "Radiation" models. Accordingly, the "Cowan" model performs much better than the "Adiabatic" one and produces very accurate results. However, the "Cowan" model does not account for the initial direct radiative heat transfer through the semitransparent material and thus overestimates thermal diffusivity (especially for coating less than three layers where a large initial direct radiative heat transfer can occur). Finally, the "Radiation + pulse correction" model of Mehling et al. [5] that accounts for the heat losses to the surroundings, the finite duration of the laser pulse and the initial direct radiation heat transfer, gives very accurate results for all coating thicknesses of 2 layers or more. It performs better than the “Adiabatic” model and the “Cowan” model, with an error that is below the standard accuracy stated by the manufacturer for the Netzsch LFA 447 (±3%).

Based on those observations, it is recommended to use the “Radiation + pulse correction” model and to apply 3 layers of graphite coating when performing LFA test on a semitransparent material.

The exact same observations and recommendations can be made for the Pyrex 7740 sample tested at 200 °C (see *Figure 5* and *Figure 6*).

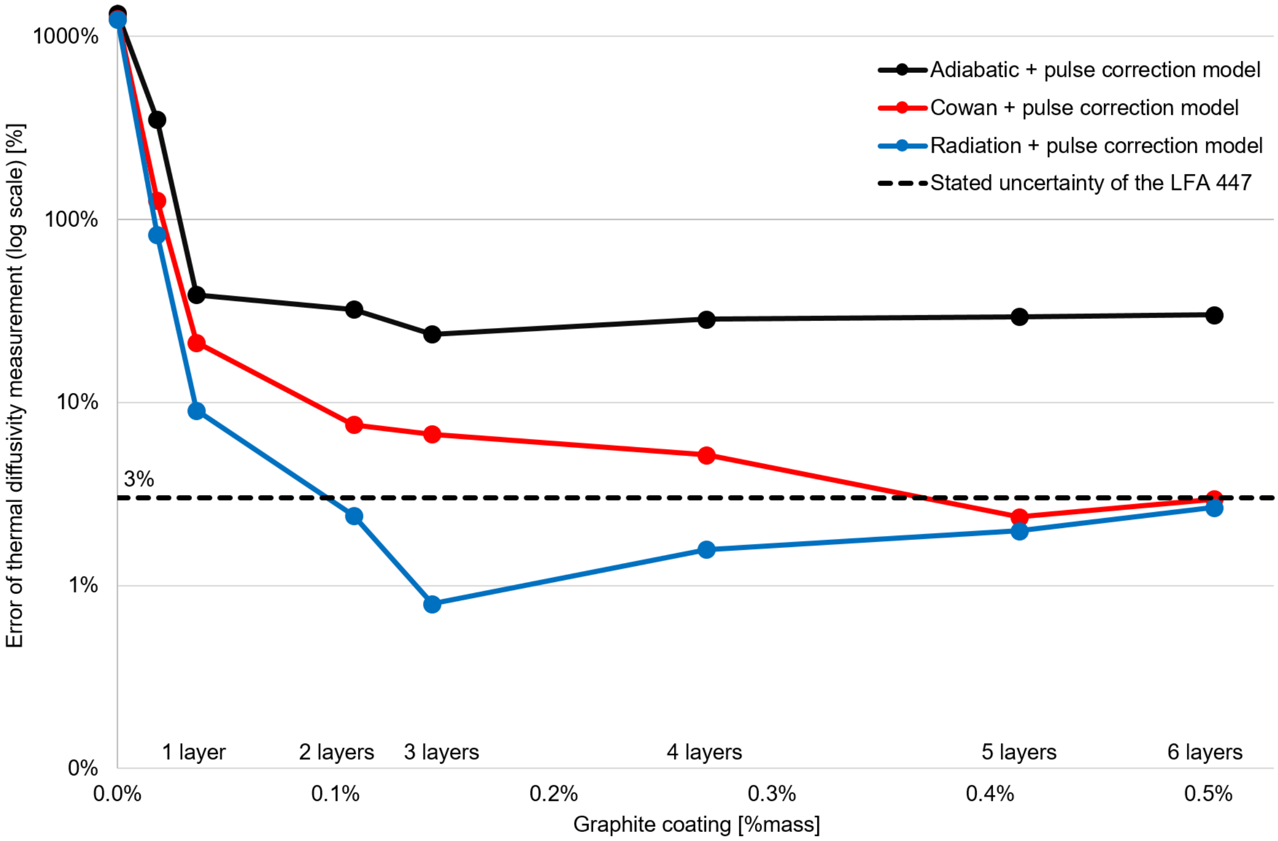

Figure 5: *Error of thermal diffusivity determination of the Pyrex 7740 (200 °C) as a function of numbers of graphite coating layers for three different fitting models.*

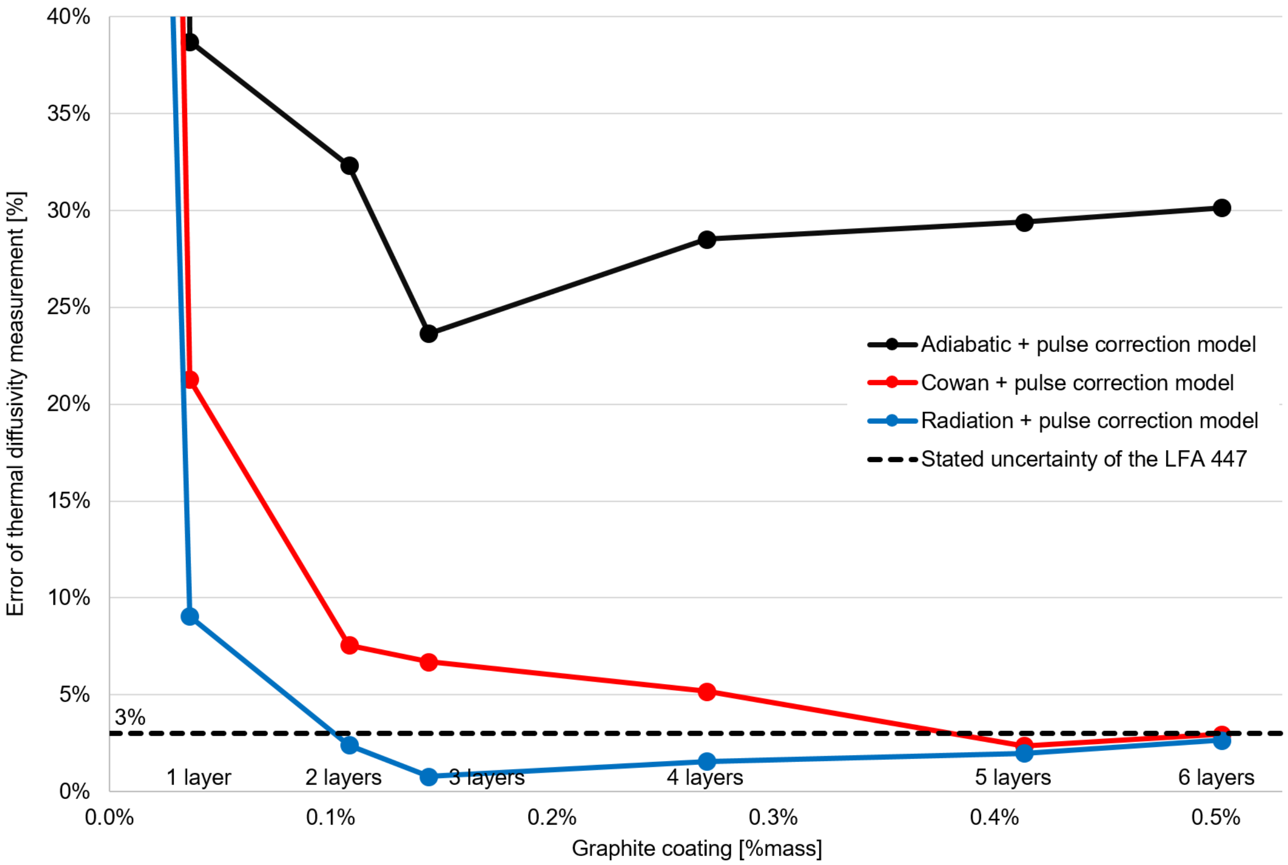

Figure 6: *Error of thermal diffusivity determination of the Pyrex 7740 (200 °C) as a function of numbers of graphite coating layers for three different fitting models.*

All these observations thus confirm the conclusions of Mehling et al. (1998) [5]. The usage of an adequate graphite coating (three layers) together with the "Radiation + pulse correction" model is sufficient to perform correct thermal diffusivity measurement by LFA method with an accuracy within the typical uncertainty range stated by the LFA apparatus manufacturer.

4.2 Pyroceram: influence of graphite coating on a gold-coated semitransparent sample

Similar tests are performed for the reference sample Pyroceram 9606 with different thicknesses of graphite coating deposited onto the original gold coating. During these tests, it was observed that an initial direct radiative heat transfer occurred on certain LFA measurements despite the gold coating (see *Figure 7*). It can thus be important to also use the “Radiation + pulse correction” for LFA measurements on gold-coated semitransparent samples when initial direct radiative heat transfer is visible on the LFA measurement signals.

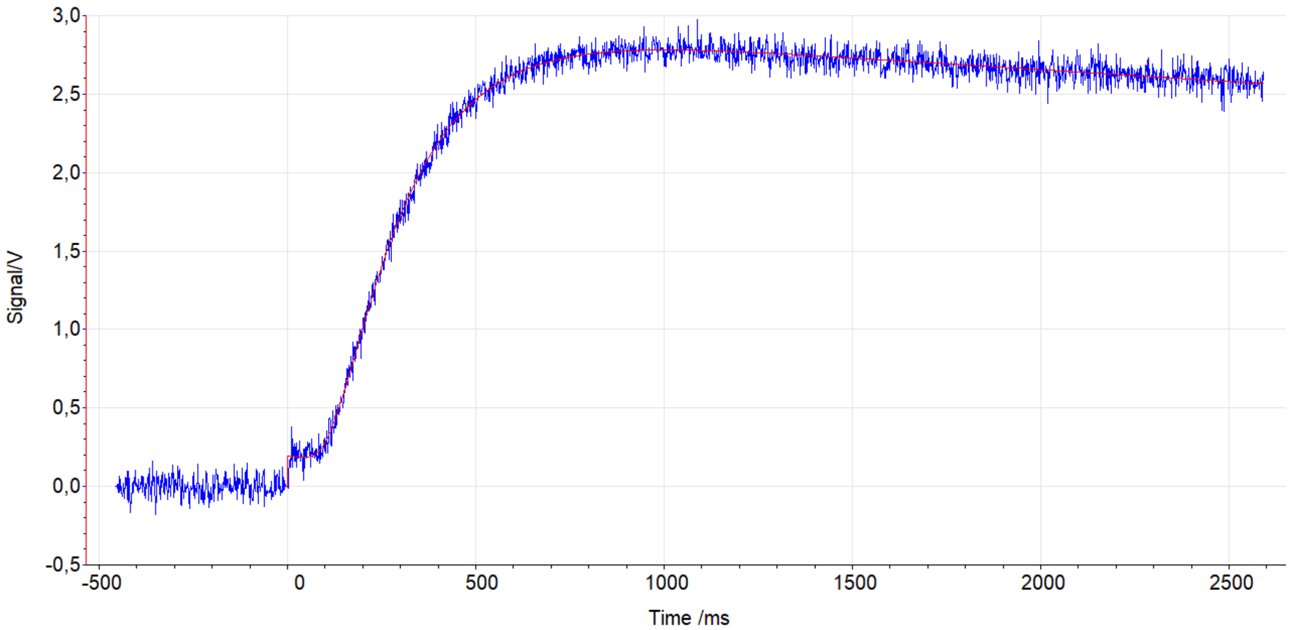

Figure 7: *An example of temperature signal from an LFA measurement performed on a gold-coated reference sample Pyroceram 9606: an initial direct radiative heat transfer is visible (initial signal step).*

One can see in *Figure 8* and *Figure 9* that the thickness of the graphite coating on the top of the gold coating does not have a significant influence on the accuracy of the thermal conductivity measurement of the LFA test, as long as there is some graphite coating. Apart from the LFA measurement at 25 °C without any graphite coating (significant overestimation of the thermal diffusivity due to improper account of the initial direct radiative heat transfer in that case), the measurement error stays the same. A large number of graphite coating layers (in here 4) does not significantly influence negatively the LFA measurement either because the additional thermal resistance of the graphite coating is negligeable in comparison to that of test sample.

Similarly to the previous test, the “Adiabatic” model overestimates the thermal diffusivity, which results in a significant error. The “Cowan” model and the “Radiation” model produce very accurate results that are mostly below the stated uncertainty threshold of 3%. However, the “Radiation” model tends to perform slightly better than the “Cowan” model. From those observations, it can be concluded that the “Radiation” model should be used for gold-coated semitransparent samples. In addition, it is recommended to spray the gold-coating with 1 or 2 thin layers of graphite coating to ensure good absorption of the laser pulse.

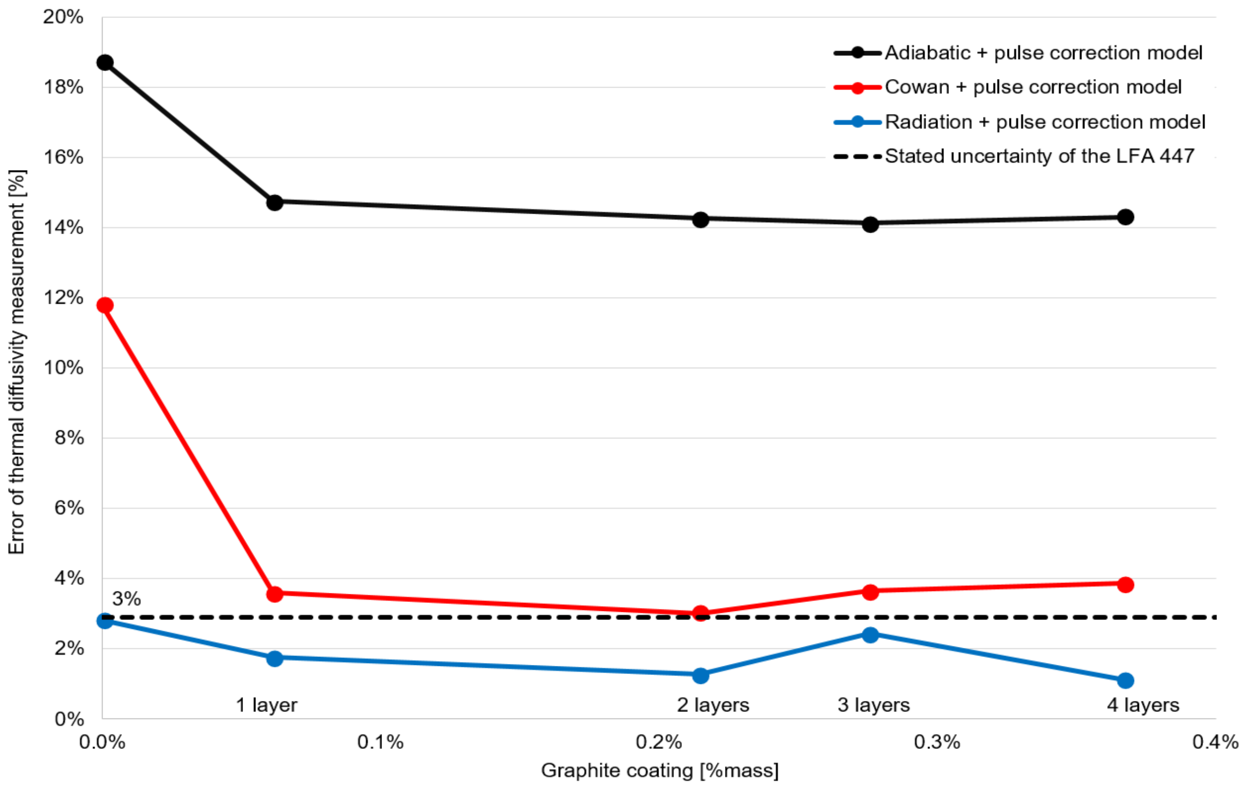

Figure 8: *Error of thermal diffusivity determination of the Pyrex 9606 (25 °C) as a function of numbers of graphite coating layers for three different fitting models.*

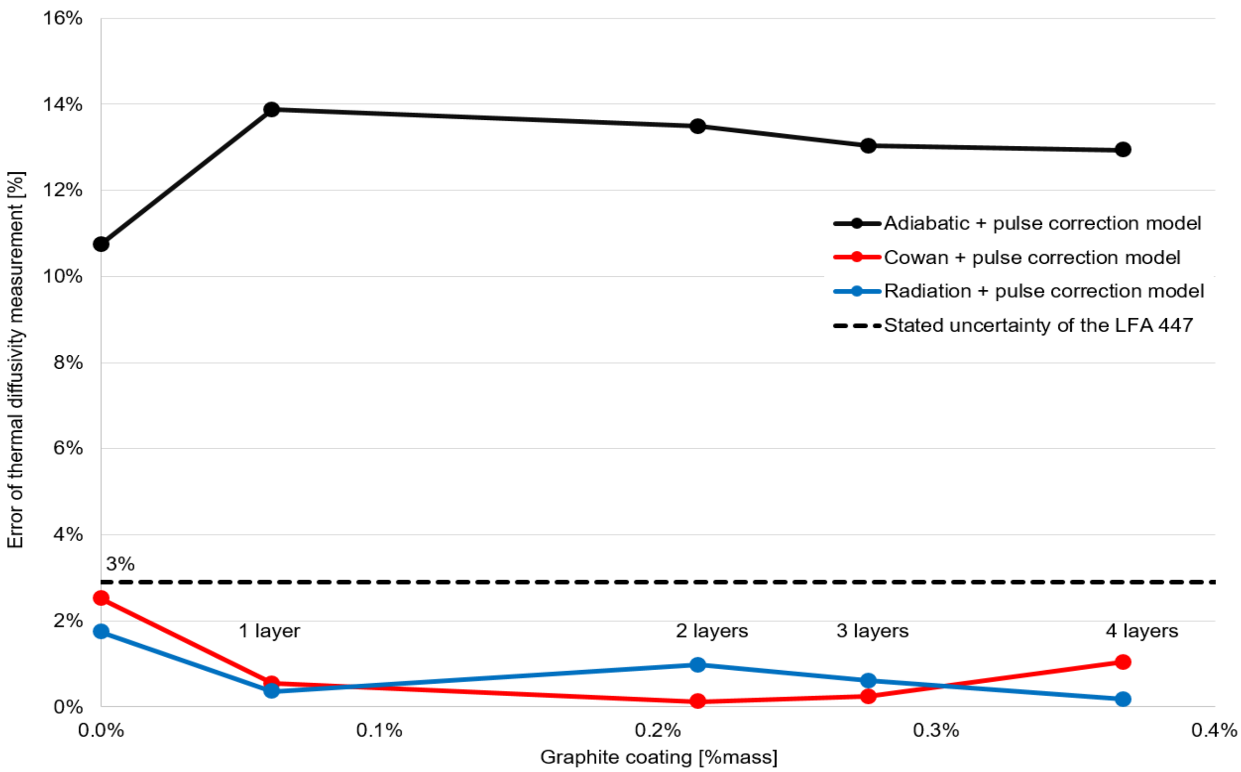

Figure 9: *Error of thermal diffusivity determination of the Pyrex 9606 (200 °C) as a function of numbers of graphite coating layers for three different fitting models.*

5. Conclusions

The observations of this experimental study confirm the conclusions of Mehling et al. (1998) [5]. The use of adequate thickness of graphite coating together with the “Radiation + pulse correction” model is sufficient to perform correct LFA thermal diffusivity measurement of semitransparent materials with “standard” thermal diffusivity (relatively low thermal diffusivity with LFA measurement duration larger than 100 ms). The measurement accuracy is thus similar to that of a gold-coated sample, and within the typical uncertainty range stated by the LFA apparatus manufacturer (below 3%).

In general, it is always recommended to coat test samples with a coating. For semitransparent materials, the gold coating is not necessary, and a graphite spray coating is sufficient. It is thus recommended to deposit 3 layers of graphite coating onto both sides of the test samples. If a gold coating is already present on the semitransparent sample, it is still recommended to apply a graphite coating of 2 layers on both sides of the sample in order to ensure laser pulse absorption and temperature surface measurement. In all cases, it is recommended to use the “Radiation + pulse correction” fitting model to accurately calculate the thermal diffusivity from the LFA test data.

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